Effect of Stochastic Fields on Spectrum of Two-level Quantum System

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***Abstract*— Analytical and numerical calculations of spectrum of two-level system, interacting with stochastic fields are presented. For the stochastic fields we consider two cases: phase-diffusion and colored-noise field of arbitrary intensity. The special attention is given to Rytov fields.**

I. Introduction And Background

STUDYING of interaction of quantum systems with various stochastic fields is of great interest both for fundamental and applied science. The stochastic fields can have both artificial and natural origin. Example of the former is laser fields. One of the most interesting examples of natural stochastic fields is the so-called Rytov fields, generated by solid surfaces. Information of Rytov field's effect on molecular characteristics near a surface is necessary for control and understanding of many processes at surface. Spectroscopic methods are the main tool in studying of interaction of quantum systems with various stochastic fields. Thus, information about line-profile, relaxation parameters and dynamics is highly required.

In the present paper we report analytical calculations of absorption spectrum and computer simulation of dynamics of two-level system, interacting with stochastic fields (phase-diffusion and colored-noise field of arbitrary intensity). The special attention is given to calculation of relaxation parameters, which contain considerable information about noise field properties. An approach to define relaxation parameters out of experimental spectrum is presented in [1].

II. Molecule-field Model

Consider two-level molecule driven by coherent probe field and interacting with stochastic field. The entire system is described by the following Hamiltonian:

$$H(t) = H_0 + \widetilde{H}(t) + V(t), \widetilde{H}(t) = d_{12}\widetilde{E}(t),\ V(t) = d_{12}E(t) \quad (1)$$

where, H_0 is the Hamiltonian for isolated system, and $\widetilde{H}(t)$ and $V(t)$ are interactions between the quantum system and stochastic and regular fields correspondingly, d_{12} is the matrix element of the electric dipole between the two states.

For analytical calculations in the case of *phase-diffusion field* (Wiener-Levy stochastic process) the equation of motion for the density matrix operator ρ with Hamiltonian (1) is written as:

$$\begin{cases} \dfrac{dN}{dt} = -2\chi(t)\,\mathrm{Im}\,\rho_{12} + \dfrac{1}{T_1}(n_0 - N) \\ \dfrac{d\rho_{12}}{dt} - i\omega_0\rho_{12} + \dfrac{\rho_{12}}{T_2} = \dfrac{i}{2}\chi(t)N \end{cases} \quad (2)$$

where n_0 is the equilibrium population difference, $N=\rho_{11}-\rho_{22}$ is the population difference, $\chi(t) = \frac{2d_{12}}{\hbar}(\varepsilon(t)e^{i\omega t} + \varepsilon^*(t)e^{-i\omega t} + E_0\cos\omega t)$, E_0 is an amplitude of the probe field and $\varepsilon(t) \equiv |\varepsilon(t)|e^{i\varphi(t)}$ is the fluctuating complex amplitude, with $|\varepsilon(t)|$ and $\varphi(t)$ being the real amplitude and phase, respectively. For calculations we consider a stationary Markov process. In this case the first-order correlation function is given by:

$$\langle \varepsilon^*(t_1)\varepsilon(t_2)\rangle = \varepsilon_0^2 \exp(-|t_1 - t_2|/\tau_c)\ ,\quad \varepsilon_0^2 = \langle |\varepsilon(t)|^2 \rangle \quad (3)$$

Analytical studies of a quantum system behavior interacting with *colored-noise field* are based on a paper [2]. In this case the Hamiltonian (1) is rewritten with Pauli matrices, r_i:

$$H_0 = -\frac{\hbar\omega_0}{2}r_3,\quad \widetilde{H}(t) = \sum_{i=0}^{3} e_i r_i \widetilde{E}(t), \quad (4)$$

$$V(t) = i\hbar\sum_{i=0}^{3}\gamma_i r_i (ae^{-i\omega t} - a^+ e^{i\omega t}),\ \gamma_i = \sqrt{\frac{2\pi\hbar\omega}{L^3}}\frac{e_i}{\hbar},$$

where r_0 is a unitary matrix and e_i are given by:

$$e_0 = \frac{d_{11} + d_{22}}{2},\ e_1 = \frac{d_{12} + d_{21}}{2},\ e_2 = \frac{d_{21} - d_{12}}{2i},\ e_3 = \frac{d_{11} - d_{22}}{2}.$$

The studies in this case are done for weakly non-Markovian noise with correlation function (3).

The *numerical calculations* (both for phase-diffusion and colored-noise fields with correlation function (3)) use equations (2) and are made for a system with parameters (in cgs): ω_0= 10.430 GHz, $d_{12}=7\cdot10^{-19}$, $n_0=3.3\cdot10^{9}$, $T_1=T_2=10^{-7}$.

In computer simulations of dynamics the Heun algorithm has been used, which allows to generally solve quantum equations of two-level system (in time-domain) even for high frequencies and intensive stochastic fields.

III. Results

1) Phase-diffusion field

The imaginary part of density matrix, ρ''_{12}, is given by ($t\to\infty$):

$$\rho''_{12} = \frac{n_0 E_0 d_{12}}{T_2^*\hbar\left((\omega - \omega_0)^2 + \dfrac{1}{T_2^*} + \dfrac{\chi_0^2 T_1}{4T_2^*}\right)},\quad \frac{1}{T_2^*} = \frac{1}{T_2} + \frac{d_{12}^2\varepsilon_0^2}{2\hbar^2(1/T_1 + 1/\tau_c)} \quad (5)$$

Thus, it is shown that the presence of *phase-diffusion field* does not affect the line profile, but the relaxation time T_2 is decreased. Dynamics of non-diagonal element of imaginary part of density matrix, ρ''_{12}, for various intensities ε_0^2 is presented on Fig.1-3 (in cgs): $\tau_c=5\cdot10^{-9}$, $E_0=5\cdot10^{-4}$.

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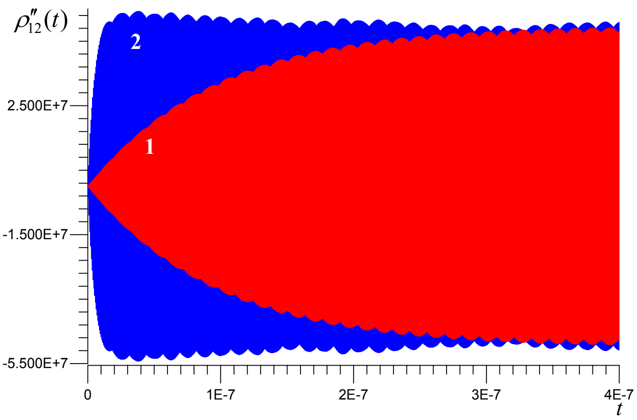

Fig.1 Computer simulation of behaviour of ρ''_{12} as a function of time: 1) ε_0^2=0, 2) $\varepsilon_0^2=10^{-4}$ (weak field), $T_2^* = T_2$ (T_2^* is calculated by (5))

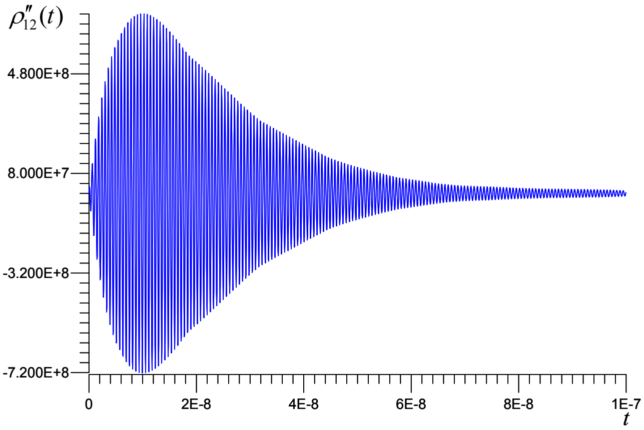

Fig.2. Computer simulation of behaviour of ρ''_{12} as a function of time: $\varepsilon_0^2=4\cdot10^{-2}$ (strong field), $T_2^*=1.7\cdot10^{-8}$.

In the short time regime, when $t<<\tau_c$, phase-diffusion field act like a coherent signal, inducing polarization in quantum system. At the moment $t\sim\tau_c$ maximum polarization is induced, and then it decays with characteristic time T_2^* defined by (5). The stationary value of ρ''_{12} is determined by (5). If $T_2^* \le \tau_c$, then polarization can be induced several times.

2) Colored-noise field

For weakly non-Markovian noise with correlation function (3) the normalized spectral line shape is given by ($\gamma\equiv1/\tau_c$):

$$I(\omega)=\frac{1}{\pi}\left\{\frac{\Omega-\omega}{\Gamma^2+(\omega-\Omega)^2}\left[\frac{(\gamma_1^2-\gamma_2^2)(\Gamma_1-\Gamma_2)+4\gamma_1\gamma_2(\Delta_1-\Delta_2)}{2\Omega}\right]+\frac{\Gamma(\gamma_1^2+\gamma_2^2)}{\Gamma^2+(\omega-\Omega)^2}\right\}$$

$$\Gamma=\frac{1}{2}(\Gamma_1+\Gamma_2)+\Gamma_3, \quad \Gamma_i=\frac{4e_i^2\varepsilon_0^2}{\gamma\hbar^2(1+\omega_0^2/\gamma^2)}+\frac{1}{2T_2}, \quad i=1,2,$$

$$\Gamma_3=\frac{4e_3^2\varepsilon_0^2}{\gamma\hbar^2}+\frac{1}{2T_2}, \quad \Omega=\left(\omega_0^2-\omega_0(\Delta_1+\Delta_2)+\Delta_1\Delta_2-\left(\frac{\Gamma_1-\Gamma_2}{2}\right)^2\right)^{1/2},$$

$$\Delta_i=-\Gamma_i\frac{\omega_0}{\gamma} \qquad (6)$$

It is seen that the line-profile is changed and central frequency is shifted. Consider a case where $d_{12}=d_{21}$, $d_{11}=d_{22}$ and $\gamma>>\omega_0$, then the profile retains Lorentzian shape and relaxation time is given by:

$$\frac{1}{T_2^*}=\frac{1}{T_2}+\frac{2d_{12}^2\varepsilon_0^2}{\gamma\hbar^2(1+\omega_0^2/\gamma^2)} \qquad (7)$$

Dynamics of ρ''_{12} for this case is presented on Fig.3 (in cgs): $\varepsilon_0^2=80$, $\gamma=1\cdot10^{12}$, $E_0=5\cdot10^{-3}$.

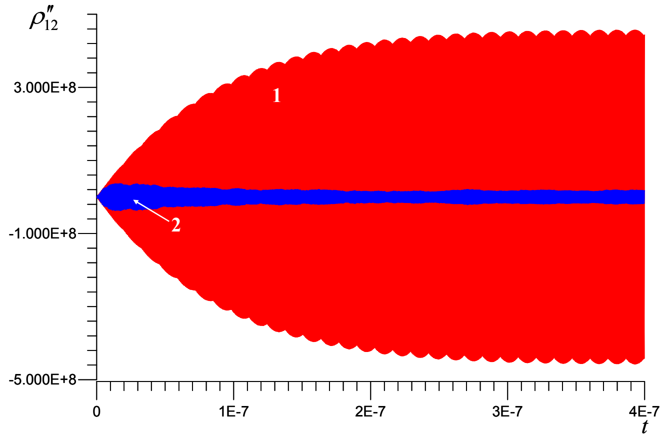

Fig.3 Computer simulation of behaviour of ρ''_{12} as a function of time: 1) ε_0^2=0, 2) ε_0^2=80 (strong field), $T_2^*=2\cdot10^{-8}$ (T_2^* is calculated by (7))

Maximal polarization is induced in $t\sim T_2^*$.

As a particular case of thermal fields, we examine the Rytov fields induced by solid surfaces. In the microwave and far IR ranges the correlation function is described by equation (3) with intensity given by (in cgs): $\varepsilon_0^2=\frac{kT}{8\pi^2h^3\sigma_0\tau_c}$, where k is the Boltzmann constant, h is a distance from a surface, σ_0 is a conductivity. Taking $\sigma_0=4.37\cdot10^{17}c^{-1}$, $\tau_c=3\cdot10^{14}$c (golden surface) and using (7), where T_2=0, we obtain: h=10A, $T_2^*=1.1\cdot10^{-6}$c; h=7A, $T_2^*=3.6\cdot10^{-7}$c; h=5A, $T_2^*=1.3\cdot10^{-7}$c. Thus, it is necessary to take into account effect of Rytov fields on an absorption line of a quantum system placed near a solid surface.

Therefore, the results of studies show that stochastic fields can strongly modify absorption line shape and dynamics of a quantum system. It is also shown that crucial role is taken by statistics properties of noise.

ACKNOWLEDGMENTS

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